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FUEL CELL
[Nenryo denchi]

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TITLE	(54):	FUEL CELL
FOREIGN TITLE	[54A]:	Nenryo denchi

Technical field

The present invention relates to a fuel cell, which is an electrochemical cell that continuously converts chemical energy produced by fuel and an oxide to electrical energy.

Background art of the technology

Generally, with fuel cells, a membrane-electrode assembly (hereafter termed MEA), wherein a macromolecular electrolytic membrane, a catalyst layer, and a reactive electrode layer are laminated between a pair of opposing collector electrodes (separators), is disposed to constitute a single cell. To generate electricity at large capacity, [fuel cells] are constituted by stacking from several tens to several hundreds of such single cells.

In fuel cells it is preferable that the dimension in the thickness orientation in particular be small in order to keep the overall size of the cell as small as possible. For this reason, it is desirable that the thickness of each component be made thin.

For the separator material, something through which current readily flows, such as carbon or metal, is selected, and from the aspect of corrosion resistance, carbon is used. The thinner the thickness, the better, and it is desirable that it be no more than about 2 mm, and preferably no more than about 1 mm. Because carbon separators of such a thickness have no elongation characteristics, they are easily broken by excessive deformation such as flexing.

* [Numbers in right margin indicate pagination of the original text.]

The positive and negative reactive electrode layers used that touch the separators are composed of porous carbon through which water and oxygen, as the fuel, can pass, and which resists corrosion. The thickness is no more than about 1 mm, preferably no more than about 500 μm, and even more preferably, no more than about 300 μm, which is thin, and because it is porous it lacks the property of withstanding deformation such as compression.

The macromolecular electrolytic membrane is an ion exchange membrane. Its thickness is no more

than about 1 mm, preferably no more than about 500 μm, and even more preferably no more than about 200 μm, which is thin. Furthermore, it is crosslinked, and because it is used in a moist state (gel state), its strength is also low.

Each component material having such a thickness that constitutes a single cell in this way has little elongation and breaks easily with deformation. For this reason, when handled roughly during assembly into cells, the components will break. When the fuel cell is constricted with strong force in order to ensure a seal, there is the risk of breakage occurring because of the weak components.

With individual unit cells constituted in this way, water vapor must be prevented from evaporating in order to keep the distance between collector electrodes constant and to control drying of the macromolecular electrolytic membrane. In the past, in order to ensure the required sealing ability relating to such prevention of drying, technology using gaskets (Japanese Kokai Patent Application Nos. Hei 7[1995]-153480, Hei 7[1995]-226220, Hei 9[1997]-231987), technology wherein a foamed sponge layer

laminated on a rubber sheet is used as a gasket (Japanese Kokai Patent Application Nos. Hei 6[1994]-96783, Hei 7[1995]-312223), and the like have been proposed.

Relating to the manufacture of single cells, it is desirable that the assembly and disassembly of cell component elements be easy, but normally such assembly and disassembly workability is sacrificed somewhat, since electricity-generating efficiency is deemed important, and a structure that is assembled [already] hardened, and fixed with adhesive, is normal.

However, even with any of the aforementioned prior art proposed to ensure sealing ability in order to prevent drying of the macromolecular electrolytic membrane, the number of manufacturing processes increases significantly, increased cost is inevitable, and it is difficult to say that characteristics satisfying the requirements are obtained over the service life of the fuel cells. In addition, while a structure wherein the individual elements of a single cell are affixed by hardened adhesion exhibit the required characteristics at the initial stage, when the cell components deteriorate with long-term use, there is the problem that replacing them is extremely difficult.

Disclosure of the invention

The objective of the present invention is to provide a fuel cell with which the desired sealing ability in a single cell is ensured and stable electricity-generating efficiency can be maintained by preventing drying of the macromolecular electrolytic membrane, and furthermore, with which replacement of deteriorated components is easy by having

excellent disassembly and assembly workability, and with which significant reduction of manufacturing costs can be realized.

The fuel cell pertaining to the present invention is such that a single cell is constituted by disposing a membrane-electrode assembly, wherein a macromolecular electrolytic membrane, a catalyst layer and a reactive electrode layer are laminated between a pair of opposing separators, and [the fuel cell] is a stack wherein multiple single cells are laminated. The membrane-electrode assembly is held between a pair of opposing gasket sheets made of resin, a gasket made of a curable rubber is formed integrally in a projecting shape on each of the outer surfaces of the gasket sheets or the inner surfaces of the separators, and the gaskets are tightly adhered to the inner surfaces of the separators or to the outer surfaces of the gasket sheets to ensure the required sealing ability.

With the constitution above, because the membrane-electrode assembly is held between a pair of gasket sheets, in the manufacture of a single cell, the membrane-electrode assembly can be positioned easily and accurately with surface pressure kept constant, and working efficiency and handling ability when a single cell is assembled are substantially improved. The required sealing ability between the separators is also ensured by the gaskets. Even when the fuel cell service period is over a long time, the seal between them is maintained, so drying caused by evaporation of water vapor in the macromolecular electrolytic membrane is prevented, and stable electricity-generating efficiency is obtained.

Brief description of the figures

Figure 1 is a longitudinal cross section showing the major portions of a single cell in a first /4

embodiment of a fuel cell pertaining to the present invention.

Figure 2 is a longitudinal cross section of the major parts of the seal of a single cell when the fuel cell is assembled.

Figure 3 is a longitudinal cross section showing the major portions of a single cell when a gasket is formed between a gasket gripping part and a spacer.

Figure 4 is a longitudinal cross section showing the major parts of a single cell when a gasket is formed on the inner surfaces of the separators.

Figure 5 is a longitudinal cross section showing the major portions of a single cell in a second embodiment.

Figure 6 shows the relative positions of the spacer and gaskets as in Figure 5, reversed.

Figure 7 shows the spacer and gasket as in Figure 6 formed on the inner surfaces of the separators.

Preferred embodiment for implementing the invention

Embodiments of fuel cells pertaining to the present invention will be explained in detail referring to the figures.

Figure 1 is a cross section showing a first embodiment of a single cell, which is the smallest unit that constitutes a fuel cell, by stacking from several tens to several hundred of them. Single cell (1) has separators (2) and (3) formed in an opposing pair of flat

rectangular shapes, and a membrane-electrode assembly (MEA), composed of a macromolecular electrolytic membrane (5) formed in the same flat rectangular shape, and reactive layers (4) and (4'), in which a catalyst layer is present on the surface or inside, are disposed between separators (2) and (3).

Holding of MEA is accomplished with macromolecular electrolytic membrane (5) in a flanged flat rectangular shape with a sufficient dimension from the outer surface. The flanged portion (5') of macromolecular catalytic membrane (5) is held along with a spacer sheet (5a) sized, for example, in the form of a frame or a long, narrow sheet, from above and below by gripping parts (6') and (7') of an opposing pair of gasket sheets (6) and (7) made of elastic resin and molded into the form of a frame by stamping, by which they are gently held. That is, the MEA is held compressed by a pair of gaskets (6) and (7) from above and below via flange part (5') of macromolecular electrolytic membrane (5) and spacer sheet (5a). In this case, flange part (5') can be held by making the spacing between gripping parts (6') and (7'), where the inside ends of gasket sheets (6) and (7) protrude, smaller than the thickness of macromolecular electrolytic membrane (5).

Gaskets (8) and (9) with a convex cross section molded using a curable rubber are integral on the outer surfaces of the outside ends of gasket sheets (6) and (7). By tightly adhering gaskets (8) and (9) to the inner surfaces (2a) and (3a) of separators (2) and (3), the sealing ability required to prevent drying of the macromolecular electrolytic membrane by evaporation of water vapor in the MEA will be ensured. The gaskets can also be formed on the inner surfaces of the separators.

As for the gaskets that are formed in a convex cross section, the width of the base part is set to about 1-3 mm, and the overall height to around 0.3-1.5 mm, for example. By making the top part of the gasket a convex cross section, the contact area with the opposing seal surface (sealed surface) will be small, and a secure seal with little constricting pressure is ensured. In this case, when the spread angle of the top part of the gasket is set to about 40-60°, sealing ability improves, so it is preferable. Because a soft material with hardness (JIS A) of no more than 60 is used, the seal surface can be sealed uniformly with good fit and little constricting pressure. Because of such secure sealing, leakage of water vapor from inside the cell and liquid outside the cell can be effectively blocked.

Here, regarding gaskets with small base width and height composed of material with low hardness, singly, the shape is not constant, handling during assembly is extremely difficult, and positioning with precision the gaskets is also extremely difficult. So, by bonding them integrally with the gasket sheets,
the soft gaskets are reinforced and handling is made easy.

By using such a gasket-gasket sheet unit, the manufacture of a single cell can be accomplished in stages. First, 2 of such gasket-gasket sheet units are positioned so that the gaskets are facing, and the aforementioned unit and the MEA are united by heat and compression bonding so that the flange part of a macromolecular electrolytic membrane is held by the gripping parts formed near the edge around the inside of the four sides of the frame-like gasket sheets. In this case, the spacer sheet could be inserted simultaneously. After this, the 2 separators above and below are stacked to form a single cell.

With a single cell manufactured in this way, the reactive electrode layer, macromolecular electrolytic sheet and the gaskets, which all have low strength, are laminated in the correct positions without suffering damage.

The fuel cell in this example using the above constitution functions as follows. The MEA is held by 2 gasket sheets (6) and (7) compression-bonded, with a macromolecular electrolytic membrane (5) and a spacer sheet (5a) between them. For this reason, the MEA can be positioned easily and accurately with constant surface pressure in the manufacture of each single cell (1), and the working efficiency and handling when single cells are assembled are substantially improved. The required sealing ability is also ensured between separators (2) and (3) by gaskets (8) and (9), and even when the fuel cell service period is long-term, the sealing ability during that period remains stable, so drying due to evaporation of water vapor in the macromolecular electrolytic membrane is prevented, and stable electricity-generating efficiency is obtained.

Because gasket sheets (6) and (7) and spacer sheet (5a) are each formed with the prescribed thicknesses, the space between the upper and lower separators is kept constant when [the components] are held between upper and lower separators (2) and (3), and the MEA, which is composed of brittle material,
is prevented from breaking.

Figure 2 shows a longitudinal cross section of the major parts of the seal of a single cell when a fuel cell is assembled. During assembly, flange part (5') of the macromolecular electrolytic membrane is compression-bonded or thermal compression-bonded to be held by end parts (6a) and (7a) (corresponding to protruding gripping parts (6') and (7')) of the gasket sheets, and the opposite surfaces between (6b-7b) and (6c-7c),

which are portions of the gasket sheets that do not deform, are also compression-bonded simultaneously. Here, polyester film, polyamide film, polyimide film, polyethylene terephthalate film or the like no thicker than about 1 mm, preferably no more than about 200 μm, and more preferably no more than about 50 μm, is used as the gasket sheet.

Next, more concrete examples will be explained.

In order to hold the MEA, a macromolecular electrolytic membrane (5) 0.2 mm thick and a spacer sheet (5a) made of stainless steel 0.7 mm thick are used, macromolecular electrolytic membrane (5) is stamped into the shape required to hold the MEA electrolytic membrane, and they are unified by thermal compression bonding.

Polyester film (Diafoil S100-100, a product of Mitsubishi Chemical Corporation) was used as the molding material for gasket sheets (6) and (7), and they were stamped into frame shapes of the required size.

In addition, gaskets (8) and (9) were molded integrally on the outer surfaces at the outside ends of gasket sheets (6) and (7) with injection molding or LIM molding. For the curable rubber, which is the material for gaskets (8) and (9), rubber that preferably adheres to thermoplastic resin film can be used. Usable the molding material for gaskets (8) and (9), is anything that is a curable rubber with hardness (JIS A) no more than 60, and preferably 20-40, and with which the required sealing ability can be ensured.

As the rubber, liquid rubber can also be used, in addition to rubber in ordinary form. As rubber in

ordinary form, for example, ethylene propylene rubber, fluorine rubber, hydrogenated nitrile rubber or other highly saturated rubber, hydrogenated styrene butadiene copolymer, hydrogenated styrene isoprene copolymer, or other highly saturated

thermoplastic elastomer or the like may be used. As liquid rubber, liquid silicone rubber, liquid nitrile rubber, liquid ethylene propylene rubber, liquid fluorine rubber or the like may be used. Because these rubbers are used united with low-strength sheet-like material such as the separators, electrodes and macromolecular electrolytic membrane, liquid rubber with low molding pressure is preferably used.

As an example when liquid rubber is used as the molding material for gaskets (8) and (9), liquid silicone rubber with viscosity before hardening of 5000-10,000 Pa/s (25°C), and hardness after hardening (JIS A) of 40 (X-34-1277A/B made by Shin-Etsu Chemical Co.) was used, the temperature of the molding die was set to 140°C and kept heated for 150 sec, and gaskets (8) and (9) were molded integrally on the surfaces of the outside ends of gasket sheets (6) and (7).

Figure 3 shows gaskets (8) and (9) formed between gasket sheet gripping parts (6') and (7') and spacer sheet (5a). In this situation, when a cell is assembled, gasket sheets (6) and (7) and separators (2) and (3) are compressed by gaskets (8) and (9) and are sealed. Thus, movement of acid toward spacer sheet (5a) from macromolecular electrolytic membrane (5) is effectively controlled, and corrosion of spacer sheet (5a) by acid is prevented.

Figure 4 shows gaskets (8) and (9) formed on the inner surfaces (2a) and (3a) of separators (2) and (3). In this case, too, corrosion of the spacer sheet caused by acid is effectively prevented.

Figure 5 is a cross section showing the major parts of a single cell (10) in a second embodiment pertaining to the present invention. Components in common with single cell (1) in the above-described first embodiment are assigned the same symbols.

In the case of single cell (10) in this example, gasket sheets (11) and (12) of different shapes are

prepared from gasket sheets (6) and (7) in the first embodiment, and spacer sheets (13) and (14) formed in an L-shaped cross section are disposed at the outer surfaces at the outside ends of gasket sheets (11) and (12), and gaskets (15) and (16) are molded integrally to hold spacer sheets (13) and (14).

A concrete example will be explained for this embodiment as well.

A macromolecular electrolytic membrane (5) 0.2 mm thick and spacer sheets (13) and (14) made of stainless steel 0.35 mm thick were used to hold the MEA indirectly, they were stamped into a rectangular or frame shape of the required size, respectively, and were united by thermal compression bonding, holding flange part (5') of macromolecular electrolytic membrane (5) between them.

Polyester film (Diafoil S100-100) was used as the molding resin material for gasket sheets (11) and (12), and it was stamped into frame shapes of the required thickness.

Additionally, at the surface near the edges around gasket sheets (11) and (12), gaskets (15) and (16) were integrally molded with the same material as that in the first embodiment.

Figure 6 shows the relative positions of spacers (13) and (14) and gaskets (15) and (16) as in Figure 5 reversed. By using such as relative position arrangement, the chance of being exposed to acid coming from macromolecular electrolytic membrane (5) is reduced, so the problem of the spacers corroding is unlikely to occur.

Figure 7 shows spacers (13) and (14) and gaskets (15) and (16) as in Figure 6 formed on inner surfaces (2a) and (3a) of separators (2) and (3).

Potential for industrial use

With the fuel cell pertaining to the present invention, a membrane-electrode assembly (MEA) is held indirectly by a pair of gasket sheets, so that the membrane-electrode assembly can be positioned easily and precisely with constant surface pressure during the manufacture of a single cell. Working efficiency and handling are substantially improved during assembly of single cells. Because replacement of deteriorated components is easy due to outstanding disassembly and assembly workability, it is economical, and there is the advantage that a significant reduction in manufacturing cost can be realized.

The required sealing ability between the separators is also ensured by the gaskets, even when the service period of the fuel cell is over a long period of time, sealing ability during that period remains stable, so that drying due to evaporation of water vapor in the macromolecular electrolytic membrane is prevented, and it is useful for obtaining stable electricity-generating efficiency.

Amendment form (Article 19)

Received by International Patent Office on January 22, 2001: initial Claim 4 in the application was withdrawn; initial Claims 1, 5 and 6 in the application were amended; the other Claims are unchanged.

(2 pages)]

Claims

1. (Amended) A fuel cell characterized in that it is a stack wherein a single cell is constituted by disposing a membrane-electrode assembly wherein a macromolecular electrolytic membrane, a catalyst layer and a reactive electrode layer are laminated between a pair of opposing separators, and wherein multiple single cells are laminated; the membrane-electrode assembly is held between a pair of opposing gasket sheets made of resin wherein a spacer sheet having a prescribed thickness is held from both sides, a gasket composed of a curable rubber is integrally molded into a convex shape on the outer surfaces of the opposing pair of gasket sheets or on the inner surfaces of the separators, and the gaskets are tightly adhered to the inner surfaces of the separators or to the outer surfaces of the gasket sheets in order to ensure the required sealing ability.

2. The fuel cell described in Claim 1 wherein a macromolecular electrolytic membrane having a prescribed thickness is furnished with a flange on the outside of the membrane-electrode assembly in order to ensure a prescribed spacing between the separators, and the membrane-electrode assembly is held by the macromolecular electrolytic membrane being held by an opposing pair of gasket sheets from both sides.

3. The fuel cell described in Claim 2 wherein the end parts of the pair of opposing gasket sheets are formed protruding so as to hold the macromolecular electrolytic membrane furnished with a flange.

4. (Removed)

5. (Amended) The fuel cell described in Claim 1 wherein the spacer sheet is positioned between gasket sheet gripping parts and gasket sheets.

6. (Amended) The fuel cell described in Claim 1 wherein a gasket is formed between a gasket sheet gripping part and a spacer sheet.

7. The fuel cell described in Claim 1 wherein the gasket is unified with the gasket sheet.

8. The fuel cell described in Claim 1 wherein a gasket molded integrally with a spacer is used.

9. The fuel cell described in Claim 1 wherein the gasket is a curable rubber with hardness (JIS A) of no more than 60.

10. The fuel cell described in Claim 9 wherein the curable rubber is a hardening material which is liquid silicone rubber, liquid nitrile rubber, liquid ethylene propylene rubber or liquid fluorine rubber.

Descriptive document based on Article 19 (1)

Claim 1 was limited to a fuel cell wherein a spacer sheet having a prescribed thickness that was stipulated in [Claim] No. 4 was held between a pair of opposing gasket sheets from both sides.

In each of the citation examples cited, there is no teaching or suggestion of a spacer sheet having a prescribed thickness being held between a pair of opposing gasket sheets from both sides.

With the fuel cell in the present invention stipulated anew in Claim 1, gasket sheets (6) and (7) and spacer sheet (5a) are each formed with prescribed thicknesses, so even when held between upper and lower separators (2) and (3), the upper and lower

separators are held, with constant spacing maintained, and the MEA, which is composed of brittle material, is prevented from breaking.

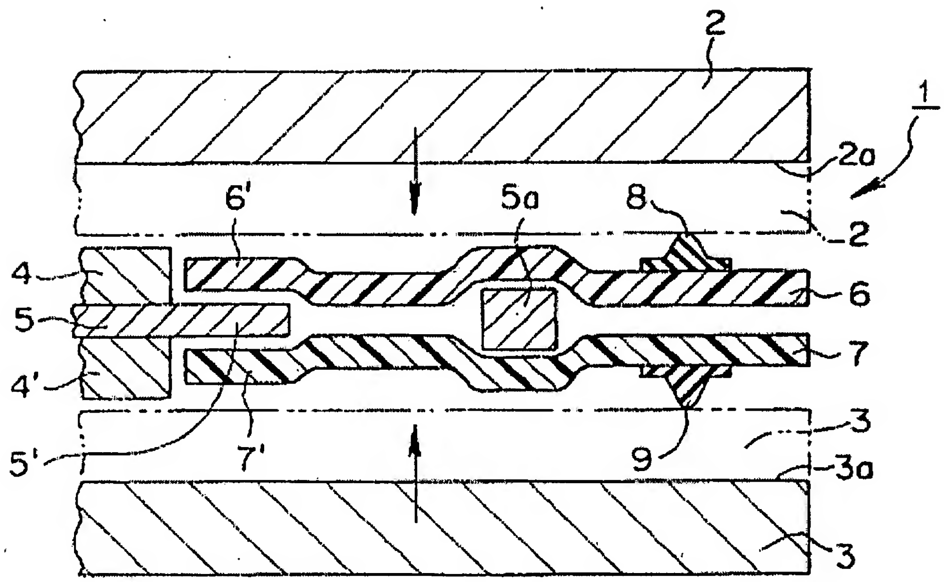

Figure 1

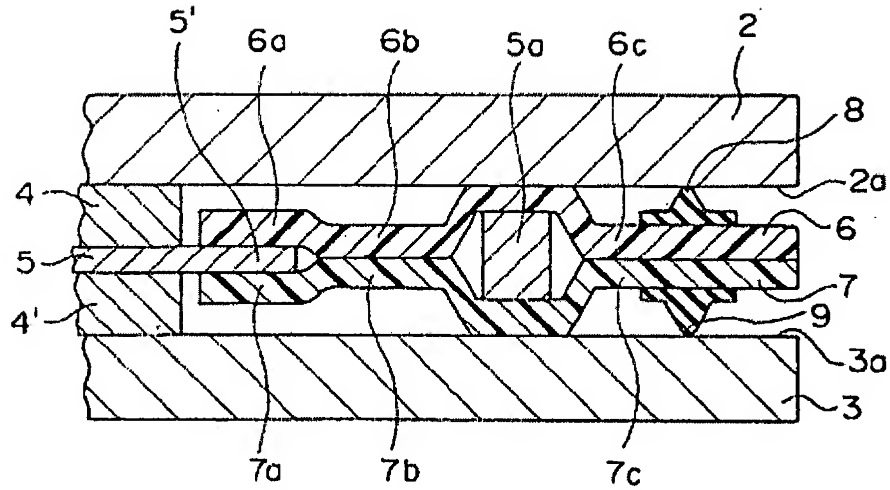

Figure 2

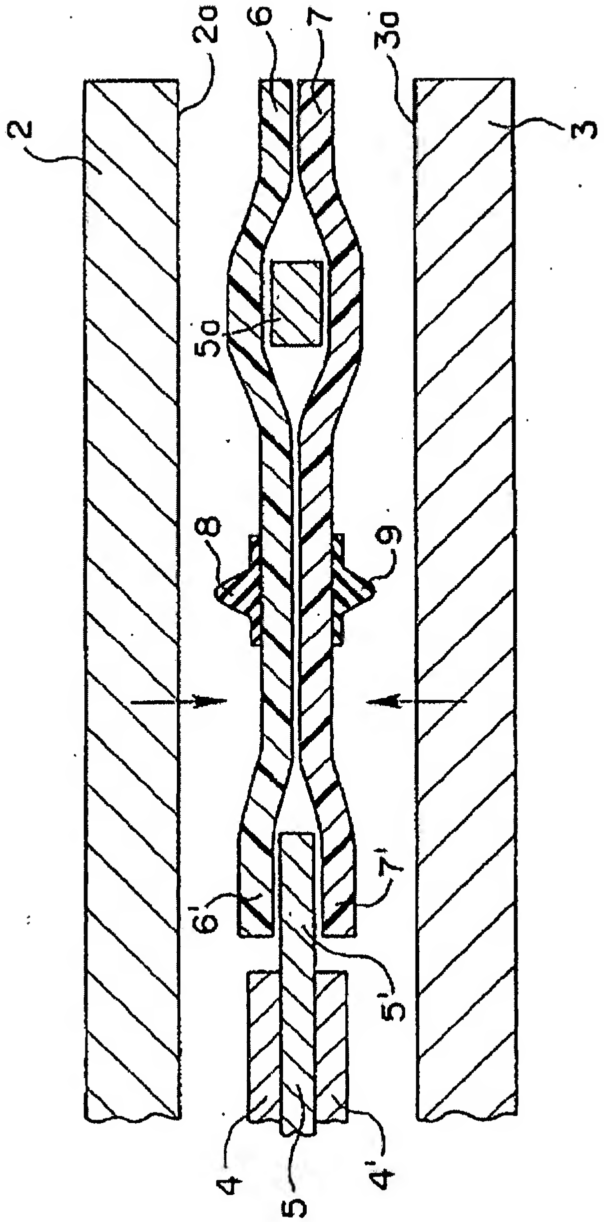

Figure 3

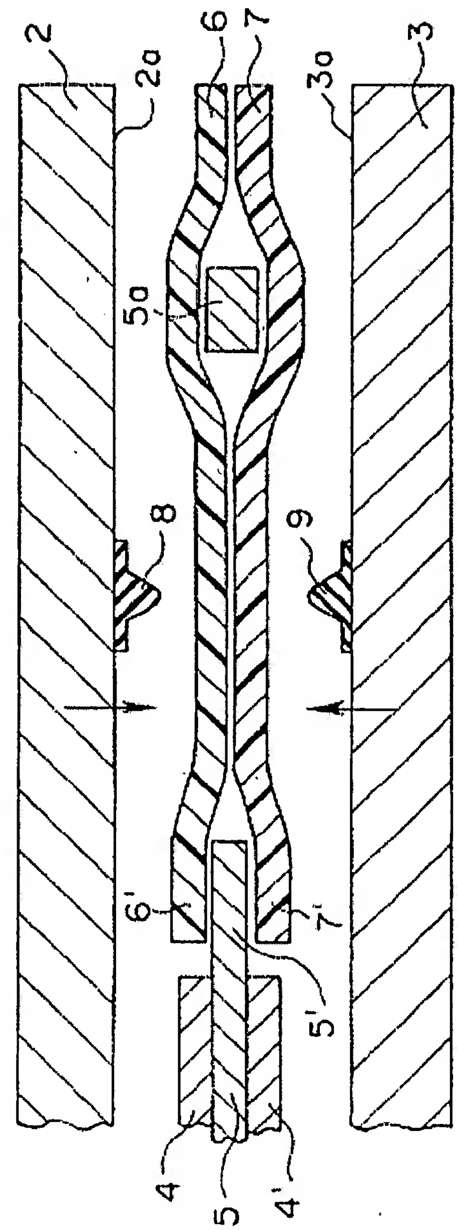

Figure 4

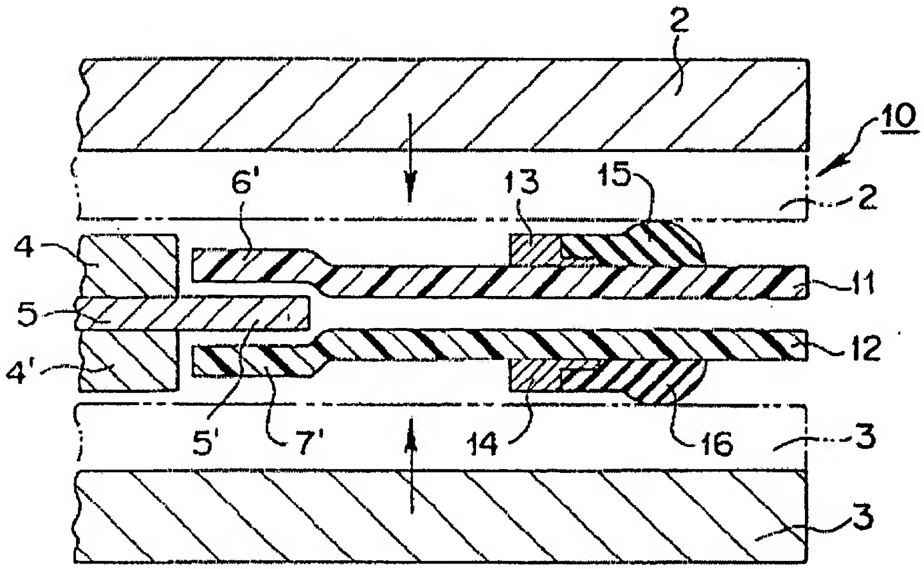

Figure 5

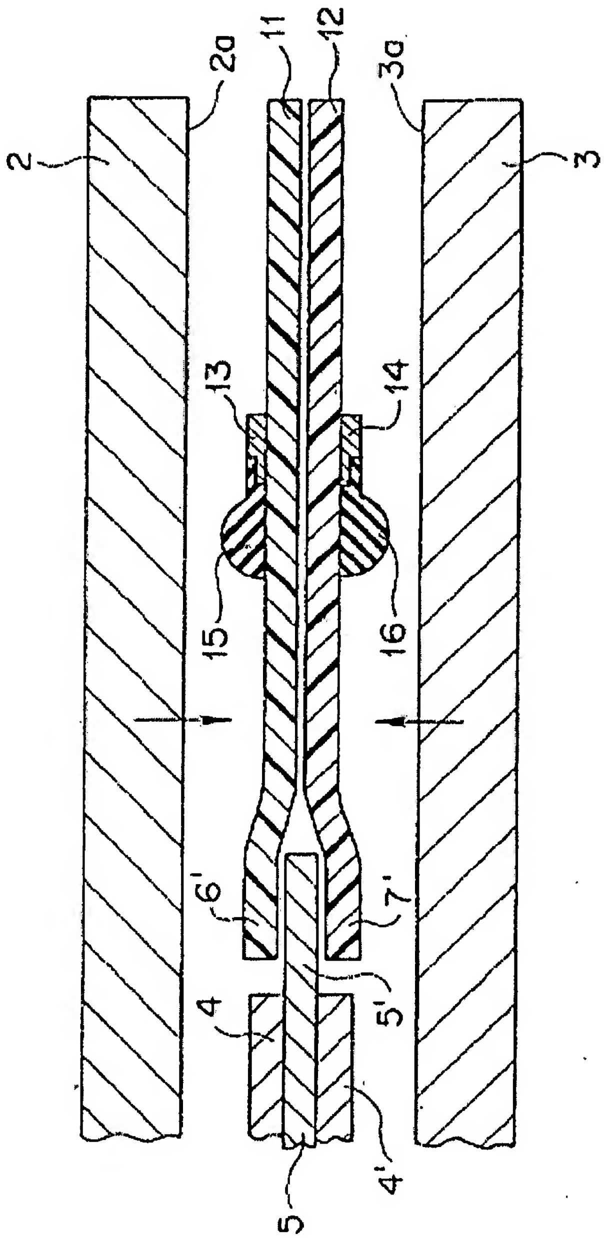

Figure 6

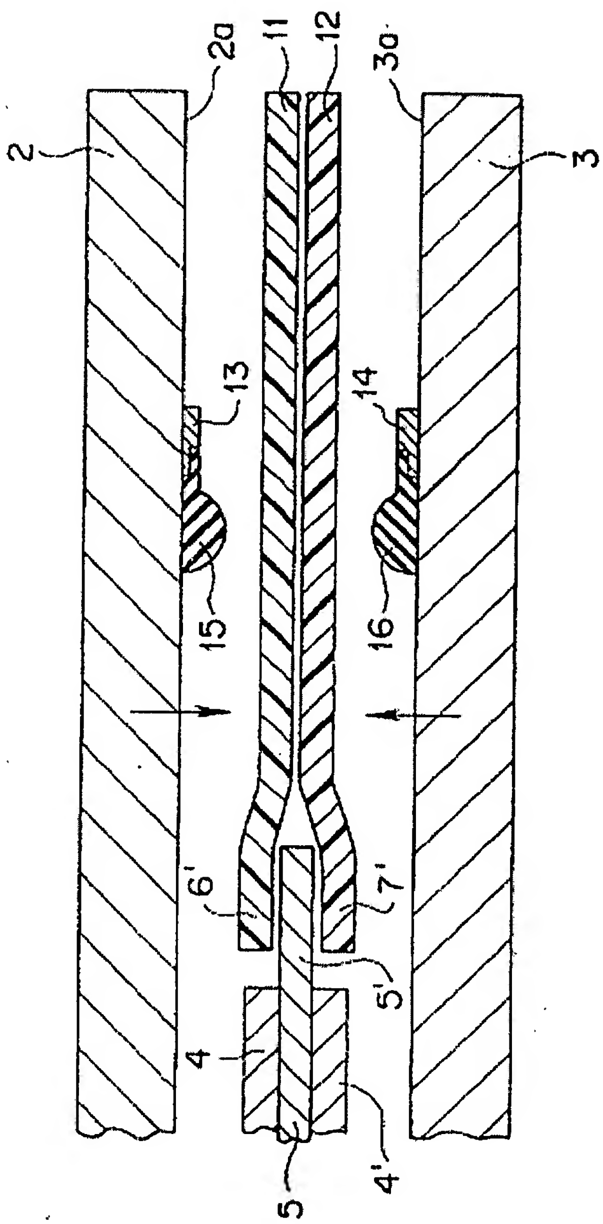

Figure 7

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP00/05855

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl[7] H01M8/02, F16J15/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
Int.Cl[7] H01M8/00-8/24, F16J15/00-15/14

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Toroku Jitsuyo Shinan Koho	1994-2000
Kokai Jitsuyo Shinan Koho	1971-2000	Jitsuyo Shinan Toroku Koho	1996-2000

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	JP, 10-55813, A (Aisin Seiki Co., Ltd.) 24 February, 1998 (24.02.98), Fig.1; page 2, right column, line 42 (Family: none)	1-3,7,9,10 1-10
X Y	EP, 914922, A1 (AISIN TAKAOKA Co., Ltd), 12 May, 1999 (12.05.99), Fig. 7 & JP, 11-129396, A	1-3,7,9,10 1-10
X Y	JP, 09-63622, A (Mitsubishi Electric Corporation), 07 March, 1997 (07.03.97), Fig. 8; page 2, left column lines 2-13; lines 44-46 (Family: none)	1-3,7 1-10
PX	JP, 2000-21422, A (Toshiba Corporation), 21 January, 2000 (21.01.00), Fig. 14 (Family: none)	1-3,7
Y	JP, 10-302814, A (AISIN TAKAOKA Co., Ltd), 13 November, 1998 (13.11.98), Fig. 2 (Spacer 9) (Family: none)	4-6,8
Y	JP, 08-279364, A (Fuji Electric Co., Ltd.), 22 October, 1996 (22.10.96), Fig. 3 (Spacer 65) (Family: none)	4-6,8

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search 21 November, 2000 (21.11.00)	Date of mailing of the international search report 05 December, 2000 (05.12.00)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 1992)